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Emergence, Complexity and Computation ECC

Andrew Adamatzky *Editor*

# Advances in Unconventional Computing

Volume 2: Prototypes, Models and  
Algorithms

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# Chapter 10

## Marangoni Flow Driven Maze Solving

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**Abstract** Algorithmic approaches to maze solving problems and finding shortest paths are generally NP-hard (Non-deterministic Polynomial-time hard) and thus, at best, computationally expensive. Unconventional computational methods, which often utilize non-local information about the geometry at hand, provide an alternative to solving such problems much more efficiently. In the past few decades several chemical, physical and other methods have been proposed to tackle this issue. In this chapter we discuss a novel chemical method for maze solving which relies on the Marangoni flow induced by a surface tension gradient due to a pH gradient imposed between the entrance and exit of the maze. The solutions of the maze problem are revealed by paths of a passive dye which is transported on the surface of the liquid in the direction of the acidic area, which is chosen to be the exit of the maze. The shortest path is visualized first, as the Marangoni flow advecting the dye particles is the most intense along the shortest path. The longer paths, which also solve the maze, emerge subsequently as they are associated with weaker branches of the chemically-induced Marangoni flow which is key to the proposed method.

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## 10.1 Introduction

Mazes and the ability to find a way through them have an intriguing and mysterious appeal to humans. They have been enshrined in the human culture for millennia, from ornaments and mythologies (e.g., the story of Theseus and the Minotaur in the Greek mythology), to contemporary fairytales, books and movies (e.g., *Maze runner* by James Dashner). The motif of a maze in human culture is unsurprisingly associated with the task of solving a complex problem with potentially many viable answers which cannot be distinguished in their entirety by a local observer. It is thus not surprising that the geometric and topological complexity of a maze and its solutions (i.e., one or more paths leading from the entrance to the exit) serves as a model configuration in many areas of science and technology (e.g., logistics, robot control, neuroscience, etc.). It has been shown that besides humans, animals, and computer algorithms, some amoeboid organisms [1–3], and even nonliving, synthetic constructs are ‘able’ to solve mazes [1–12]. Such chemical, physical or biological systems are initially in a non-equilibrium thermodynamic state with a spatial gradient of some thermodynamic variable, e.g., temperature, chemical potential, pressure, electric or magnetic field, which induces a flow of matter (momentum) or energy within the system to reach its equilibrium state. Some of the most prominent approaches are briefly mentioned next. Microfluidic networks are often solved by imposing a pressure gradient across the corresponding maze [4] between the entrance and the exit so that the pressure-induced flow has the largest amplitude along the shortest path. An electric field gradient was used to induce a glow discharge in gas-filled microchannels and to identify the shortest path in mazes or urban city maps [5, 6]; in a medium conducting electric current the shortest path is characterized by the largest gradient of the electric field which ionizes a gas and induces a plasma glow. Maze solving by a network of memristors is also based on the presence of an electric potential gradient [7]. Chemical and electric potential wave propagation along a dendritic tube of a single cell organism is the most commonly employed setup for identifying the shortest path between two food sources in a biological system [1–3]. Finally, in chemical systems a chemical potential gradient created at the beginning of the experiment induces a flow of matter which highlights the shortest path in a maze in a number of distinct ways [8–13]. A silver ion gradient initiates the propagation of a chemical wave in the Belousov–Zhabotinsky solution along the paths of a maze with the fastest wave corresponding to the shortest path [8, 9]. The concentration gradient of sodium acetate initiates the propagation of a supersaturation front in a complex structure of a hot ice computer [10]. A pH gradient is responsible for the movement of a surfactant covered organic droplet in a maze filled with an alkaline solution [11]. When a surfactant is in the system, the formation of a pH gradient has more intricate consequences on the resulting macroscopic dynamics in the maze. Surfactants reduce surface tension and the concentration of the fatty acid surfactant depends on the pH of the medium. Therefore, the pH gradient creates a difference between the surface tension of the two sides of the droplet facing the acid and base, making it move in the direction of acid.

In this chapter we show that the so called Marangoni flow induced by a pH gradient can be used as an operator for efficient maze solving. For the practical realization of such a chemical computer it is necessary to mention that we have to deal with a liquid chemistry environment [12, 14].

## 10.2 Experimental

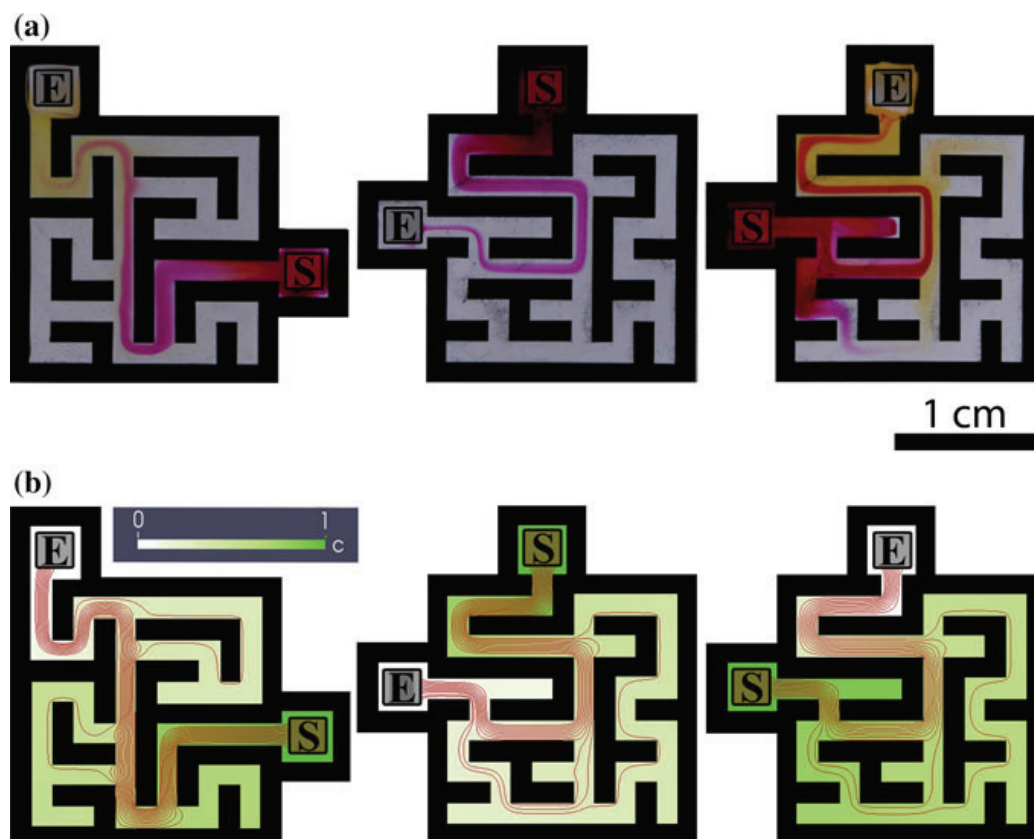
First, mazes with various topological complexity and spatial extent were designed and fabricated from polydimethylsiloxane (PDMS) using photo- and laserlithography (with thickness and depth of 1.4 and 1 mm, respectively). In a typical experiment the maze was filled with a 0.05 M alkaline solution of potassium hydroxide (KOH, Sigma-Aldrich) containing 0.2 % of 2-Hexyldecanoic acid (Sigma Aldrich) (2-HDA). 2-HDA is a fatty acid and by itself is not soluble in water. However, in alkaline solution the head group is deprotonated and becomes soluble. Consequently, the fatty acid molecules are oriented at the liquid-air interface and the deprotonated form of 2-HDA acts as surfactant (reducing the surface tension at the liquid-air interface). An acidic hydrogel (Agarose, Sigma-Aldrich) block ( $\sim 1 \times 1 \times 1$  mm) was placed at the exit of the maze. After addition of an acidic block, a small amount ( $\sim 0.3$  mg) of dry Phenol Red dye powder was placed at the liquid-air interface at the starting point (the other entrance of the maze). With this technical set-up of the maze and the necessary chemical reaction and diffusion partners, we can run a time resolved experiment where we observe and track the spatial transient of the colorization of the paths through the maze.

## 10.3 Results and Discussion

In the presented experimental setup the dye particles traveled passively at the liquid-air interface towards the acidic hydrogel block, i.e., the region of low pH. The dye particles transported by the pH-induced Marangoni flow gradually dissolved in the water phase and the color showed their paths through the maze. Figure 10.1a shows maze solving experiments in various mazes filled with an alkaline solution of a fatty acid. The symmetry in the system is broken by an acidic hydrogel. In a typical experiment, the shortest path can be found and visualized within  $\sim 10$  s (Fig. 10.1a).

The Marangoni flow facilitating the maze solving is induced by the non-uniform distribution of the surface tension at the liquid-air interface, and it drives transport of the top fluid layer towards the higher surface tension regions from the low surface tension regions. The intensity of the fluid flow is propositional to the gradient of the surface tension. Addition of an acidic block to the maze filled with an alkaline solution of a fatty acid changes the surface tension of the solution at the liquid-air interface. This surface tension difference (surface tension gradient) creates and maintains a fluid flow in the liquid phase which is commonly referred to as the Marangoni flow.

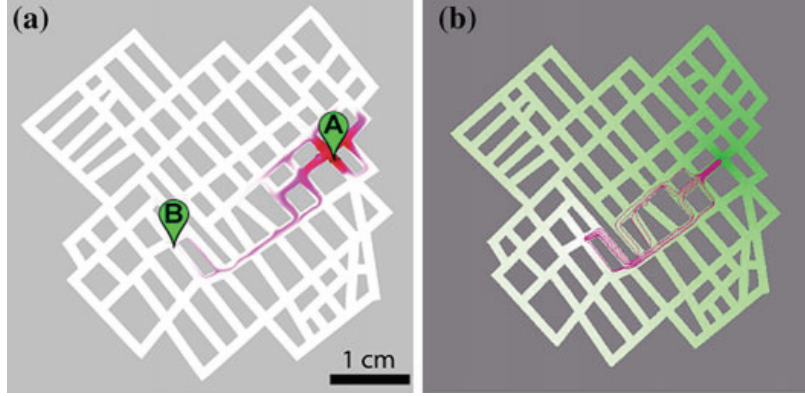




**Fig. 10.1** Maze solving and finding the shortest path in various mazes, experiments (a) and numerical simulations (b). Position of the gel soaked with acid (exit) is indicated by letter *E*. Letter *S* shows the entrances of the maze (starting point), where Phenol Red dye particles (passive tracer) are added. Reprinted from [12] under Creative Commons Attribution 4.0 International Public License

The most intense fluid flow is established along the shortest path in a maze, where the gradient of the surface tension at the liquid-air interface is the highest. Thus, the most of the tracer particles are caught up in the dominant flow branch along the shortest path which is thus characterized by the most intense color contrast of the dissolved dye. However, it should be noted that in a relatively complex maze all possible (and not only the shortest) solution paths can be explored by the Marangoni flow provided that a sufficiently long time is allowed. In our setup it is  $\sim 60$  s instead of 10 s which was sufficient for exploring just one path (the shortest path).

The existence of the gradient of the surface tension in a maze can be explained by the effect of pH on the protonation rate of fatty acid molecules. Fatty acid molecules can be protonated/deprotonated by different extent depending on the pH of the medium. Therefore, pH can be used as a technical control parameter to create and maintain a Marangoni flow in a channel network. We performed a range of numerical simulations in mazes to verify the experimental concept and obtain the shortest paths (Fig. 10.1b). Figure 10.2 shows the numerically simulated analogue of the determination of the shortest path between two arbitrary points in a complex maze (downtown

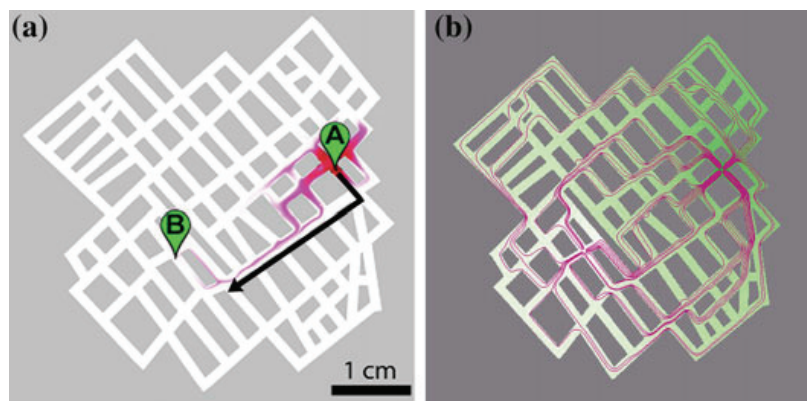


**Fig. 10.2** Finding the shortest path between two points in a channel network (made from PDMS) based on the street map of downtown of Budapest, experiments (a) and numerical simulations showing major streamlines (b). Position of the gel soaked with acid (end point) is indicated by letter *B*. Letter *A* shows the starting point, where Phenol Red dye particles are added. Figure 10.2a is reprinted from [12] under Creative Commons Attribution 4.0 International Public License

of Budapest in this case). The streamlines associated with the possible paths were calculated from the gradient of a stationary concentration  $c(x, y)$  between two points (marked by letter *A* and *B* in Fig. 10.2) which is calculated from the time-invariant solutions of the diffusion equation in two-dimensions given by

$$\frac{\partial c}{\partial t} = D \left( \frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right), \quad (10.1)$$

with Dirichlet boundary conditions,  $c = 1$  at *A*, and  $c = 0$  at *B*. The solutions procedure involved the ADI (Alternating Direction Implicit) method to solve (10.1) on a  $1000 \times 1000$  uniform rectangular grid. Finally, the streamlines were calculated from the gradient field of the concentration. The simulation results capture the main features of the experiments. Importantly, the shortest and the second-shortest paths found in the chemical computer experiments are in good agreement with the paths detected in the numerical simulations. Several other solution paths visible in the simulations are less pronounced in the experiments. This discrepancy could arise from the finite viscosity of the fluid in the maze (the dynamics of the fluid is not yet considered in the purely diffusive simulations of (10.1)) and the finite time duration of the experiment. In experiments with channels where the surface tension gradient is weak enough, the fluid viscosity suppresses the generation of the convective flow. Moreover, the elapsed time in the real experiment is finite, whereas the numerical solution represents the stationary, time-asymptotic solution. One additional aspect pertaining to this method deserves a mention. Theoretically, the path that connects the bottom-right branch of the site *A* to *B* (see arrow in Fig. 10.3) should be the third-shortest path, although it is not found and observed neither experimentally or numerically.



**Fig. 10.3** Finding the shortest path between two points in a channel network based on the street map of downtown of Budapest, experiments (a) and numerical simulations showing all streamlines (b). Figure 10.3a is modified from [12] under Creative Commons Attribution 4.0 International Public License

Hence, the geometrically-third-shortest path is not detected in this system. It would require more conceptual and experimental efforts to raise this kind of orthogonal solution as well. Interestingly, this property is both disadvantageous and advantageous. The path-finding method presented here does not guarantee identification of geometrically shortest paths, since it is based on the real physicochemical state of the system. Consequently, the experimental outcome could be disturbed by the fluid resistance, environmental perturbations and the system size, etc. On the other hand, the system can be treated as a solver that handles the path-finding problem with realistic constraints, which might be very inefficient to solve ‘on-the-fly’ algorithmically. The presented method allows for detecting those solution paths which are the physicochemically-permitted shortest ones, including the restriction of the channel capacity, rather than the geometrically-optimal paths. This property would be important when we consider an application, for example the chemically-induced transport of chemical entities (e.g., drug delivery).

## 10.4 Conclusions

In this chapter we presented a novel approach to maze solving which is achieved by generating a pH induced Marangoni flow in the considered channel network. Our method is based on the simple fact that pH change can affect the surface tension of a fatty acid solution, and it generates a surface tension difference at the liquid-air interface. The resulting global Marangoni flow can transport passive soluble dye particles from the starting point (high pH region) to the exit of the maze (low pH region) and in the process highlight the possible solution paths to the maze problem. It is worth pointing out that other non-local phenomena affecting the surface tension of liquids or solutions can be also utilized for maze solving (e.g., temperature, irradiation). These might represent for some communities the technically more welcome control mechanisms in practice depending on the experimental constraints.



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